NATURAL α ACTIVITY IN PARTICULATE MATTER PM2.5

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Abstract. Several open-pit coal mining areas are sited close to Forschungszentrum Jülich, Germany. There was public concern about the respirable fraction of particulate matter PM2.5 resulting from the mining activities. It was suspected that this fraction is loaded with enhanced α activity, especially ^{210}Po, which originates from ^{222}Rn decay. This would lead to increased lung exposure to radioactivity by respirated particulate matter for individuals living in the Jülich area. Therefore, since October 2009, Forschungszentrum Jülich has been monitoring the α activity of PM2.5 at two different heights (30m and 120m) at its meteorological tower. The α activity of the filters with an exposure time of one week is determined every six months. From the increase in α activity, which is due to the ^{210}Pb decay, the initial ^{210}Pb activity can be calculated. Because of their short half-lives, the nuclides of the decay chain between ^{222}Rn and ^{210}Pb are not taken into account. Knowing the air flow rate of the PM2.5 sampling unit, an annual inhalation dose is calculated based on the derived ^{210}Pb activity. The resulting annual effective dose for children <= 1 year is 8.7 ± 4.8 µSv and for adults 10.5 ± 5.8 µSv. The respective lung dose for children <= 1 year is 72.5 ± 40.2 µSv and for adults 85.9 ± 47.6 µSv.

1. INTRODUCTION

Forschungszentrum Jülich, Germany, is surrounded by several open-pit brown coal mining areas. Measurements by the State Office for Environmental Protection in North Rhine-Westphalia (LANUV) showed that the European limit on the number of days on which the PM10 value of 50 µg per square meter may be exceeded was frequently violated in the last few years in the Jülich region. With respect to these findings, there was public concern about whether the particulate matter emissions resulted from the mining activities, and whether brown coal particulate matter transported from the open pit by the wind was loaded with a higher specific natural α activity than house dust and would therefore lead to a higher lung exposure than in areas with lower PM exposure.

The Department of Safety and Radiation Protection of Forschungszentrum Jülich performs a routine environmental monitoring program related to artificial radioactivity in the vicinity of the Forschungszentrum. Beginning in October 2009, this monitoring program was temporarily extended to include a particulate matter PM2.5 sampling unit at the meteorological tower on the Jülich campus at a height of 120m. A second PM2.5 sampling unit was set up at a height of 30m in March 2010. Both sampling units are still in operation. Because PM2.5 is the respirable fraction of the particulate matter, an inhalation dose for an individual with respect to natural dust exposure can be derived from the monitoring program.

2. SETUP

Progeny from the ^{238}U decay chain up to ^{226}Ra are fixed, mainly in the soil. ^{226}Ra decays to ^{222}Rn, which is passed from the soil into the atmosphere. The mean lifetime of ^{222}Rn is 4.77E+05 sec. The following nuclides in the decay chain up to ^{210}Pb have mean halftime of the order of seconds or minutes. A very short time after decay a ^{222}Rn atom has therefore been converted into ^{210}Pb, which is a low-energy β emitter. The mean lifetime of ^{210}Pb (1.0E+09 sec) is very long compared to that of all the nuclides resulting from ^{222}Rn decay. The ^{210}Pb decays to ^{210}Po, which is an α-emitter with a mean lifetime of 1.72E+07 sec. Since the short-lived precursors of ^{210}Pb, especially ^{218}Po, tend to be attached to atmospheric aerosols [1], [2], ^{210}Pb can be considered a base product for a potential lung dose due to the inhalation of the aerosols by an individual. The dose mainly results from the α-decay of the daughter nuclide ^{210}Po.

^{220}Rn also escapes also from the soil from the ^{232}Th decay chain. A typical range of concentration in the atmosphere is 0.01 – 1 Bq m^{-3}, which is about a factor of 100 lower than that for ^{222}Rn [3]. The mean lifetimes of the decay products of ^{220}Rn are rather short (max. 5.53E+04 sec for ^{212}Pb). It is reported [4] that ^{212}Pb also tends to attach to aerosols. Considering atmospheric aerosols as a source of a potential lung dose, this decay chain is of minor importance since the dose conversion factor of ^{212}Pb is more than one order of magnitude lower than that of ^{210}Pb [5]. In [6] the authors state that the annual inhalation dose resulting from the ^{232}Th decay chain in Canada is about one order of magnitude lower than the respective dose from the ^{238}U chain.

Due to its very low energy ($\overline{E_1}$ = 4.1 keV, $\overline{E_2}$ =16.1 keV) the β emission of ^{210}Pb is not normally detected by the laboratory equipment in the routine monitoring program. However, the inhalation dose is mainly determined by the initial ^{210}Pb load of the aerosol. Therefore, one week after the end of the PM2.5 sampling period, the filters were analyzed with respect to their α activity, which is thought to result from the decay of ^{210}Po, which in turn is a decay product of ^{210}Pb. Observing the non-linear growth of ^{210}Po α activity for a period of at least 12 months by one measurement every six months allows a conclusion to be drawn concerning the initial ^{210}Pb load on the filter.

3. THEORY

The relation between the activity A and the number of decaying nuclei N is given by

$$A = \lambda N \tag{1}$$

with λ being the decay constant.

The actual number of ^{210}Pb nuclei at time t is determined by the decay law

$$N_{^{210}Pb}(t) = N_{^{210}Pb_0} e^{(-\lambda_{^{210}Pb} t)} \tag{2}$$

where $N_{^{210}Pb_0}$ is the initial number of ^{210}Pb nuclei at $t = 0$.

The growth in the number of ^{210}Po nuclei is derived from the decay law.

$$N_{^{210}Po}(t) = N_{^{210}Pb_0} \left(\frac{\lambda_{^{210}Pb}}{\lambda_{^{210}Po} - \lambda_{^{210}Pb}}\right) \left[\exp(-\lambda_{^{210}Pb} t) - \exp(-\lambda_{^{210}Po} t)\right] \tag{4}$$

Evaluating Eq. (4) with respect to the results of the α activity measurements of the filters, in addition to the initial number of ^{210}Pb nuclei, the effective start of decay is also unknown.

In this investigation Eq. (4) will be expressed in the following form:

$$N_{^{210}Po}(t - t_0) = N_{^{210}Pb_0} \left(\frac{\lambda_{^{210}Pb}}{\lambda_{^{210}Po} - \lambda_{^{210}Pb}}\right) \left[\exp(-\lambda_{^{210}Pb}(t - t_0)) - \exp(-\lambda_{^{210}Po}(t - t_0))\right] \tag{5}$$

with t being the time after the end of sampling and t_0 being the unknown start of decay.

Rearranging Eq. (5) yields

$$N_{^{210}Po}(t - t_0) = a\,L\exp(-\lambda_{^{210}Pb} t) - b\,L\exp(-\lambda_{^{210}Po} t) \tag{6}$$

with $a = N_{^{210}Pb_0} \exp(\lambda_{^{210}Pb} t_0)$ and $b = N_{^{210}Pb_0} \exp(\lambda_{^{210}Po} t_0)$,

$$L = \frac{\lambda_{^{210}Pb}}{\lambda_{^{210}Po} - \lambda_{^{210}Pb}}$$

The parameters a and b in Eq. (6) are fitted from the α activity measurement results using the Levenberg-Marquardt algorithm [7]. The unknown numbers of ^{210}Pb nuclei and the unknown start of decay (t_0) are derived by solving the linear equation system

$$\begin{pmatrix} 1 & \lambda_{^{210}Pb} \\ 1 & \lambda_{^{210}Pb} \end{pmatrix} \begin{pmatrix} \ln(N_{^{210}Pb_0}) \\ t_0 \end{pmatrix} = \begin{pmatrix} \ln(a) \\ \ln(b) \end{pmatrix} \quad (7)$$

Applying Eq. (1) yields the initial ^{210}Pb activity $A_{^{210}Pb_0}$.

The inhalation dose is calculated by

$$D_i = A_{^{210}Pb} \frac{1}{AF} BR T_e DCF \quad (8)$$

with $A_{^{210}Pb}$ = ^{210}Pb activity [Bq], AF = air flow through the sampling unit [m^3], BR = breathing rate [m^3 s^{-1}], T_e = exposure time [s], and DCF = dose conversion factor [Sv Bq^{-1}].

Table 1. Parameters used for dose calculation

Parameter unit	BR m^3 s^{-1}	T_e s	DCF_{Lung} Sv Bq^{-1}	$DCF_{Effective}$ Sv Bq^{-1}
Child <= 1 year	6.05E-05	3.15E+07	1.5E-04	1.8E-5
Adult	2.33E-04	3.15E+07	4.6E-05	5.6E-6

4. RESULTS AND DISCUSSION

Applying the Levenberg-Marquardt algorithm to the results of the α measurements and taking the air flow through the sampling units into account yields the resulting initial ^{210}Pb concentration and the t_0-parameters which are listed in Table 2.

Table 2. Resulting ^{210}Pb concentration and t_0 values (cf. Eq. (5))

Filter	Start of Sampling	End of Sampling	^{210}Pb [µBq m^{-3}]	t_0 d
120/1	21/10/2009	28/10/2009	363.9	-6.7
120/2	28/10/2009	04/11/2009	316.8	-9.6
120/3	04/11/2009	11/11/2009	105.1	-20.0
120/4	11/11/2009	19/11/2009	242.3	-5.4
120/5	19/11/2009	25/11/2009	580.0	-8.7
120/6	25/11/2009	02/12/2009	71.0	-25.9
120/7	02/12/2009	09/12//2009	119.2	-17.6
120/8	09/12/2009	16/12/2009	270.2	-4.2
120/9	16/12/2009	23/12/2009	357.1	-13.0

120/10	06/01//2010	14/01/2010	519.5	-5.4
120/11	14/01/2010	20/01/2010	227.0	-8.4
120/12	20/01/2010	27/01/2010	499.2	-5.9
120/14	03/02/2010	10/02/2010	391.0	-6.1
120/15	10/02/2010	17/02/2010	469.2	-8.3
120/16	17/02/2010	24/02/2010	204.3	-8.6
120/17	24/02/2010	03/03/2010	98.0	-25.0
120/18	03/03/2010	10/03/2010	151.2	-14.0
120/19	10/03/2010	17/03/2010	89.3	-57.6
120/20	17/03/2010	24/03/2010	231.4	-15.7
120/21	24/03/2010	31/03/2010	180.4	-16.6
120/22	31/03/2010	07/04/2010	139.4	-19.3
120/23	07/04/2010	14/04/2010	279.3	-19.7
120/24	14/04/2010	21/04/2010	224.7	-26.7
120/25	21/04/2010	28/04/2010	206.1	-32.5
120/26	28/04/2010	05/05/2010	197.8	-22.8
120/27	05/05/2010	12/05/2010	96,4	-45,7
Mean			247.9	-18.0
Standard Deviation			144.6	12.9

Of course, the ^{210}Pb concentration and t_0 values given in Table 2 are fictitious since the adsorption of ^{210}Pb on aerosols is a continuous process and does not occur suddenly. However at the end, about 95% of ^{210}Pb is adsorbed [2]. Gründel and Porstendörfer [2] report that for long-lived radon decay products the activity size distribution changes to greater particle diameters (~550 nm). This is mainly due to coagulation processes with non-active aerosol particles during their residence time in the atmosphere. The longer residence time in the atmosphere is also reflected in the t_0 values [cf. Table 2], which mostly go back to the time before sampling started before sampling started (i.e. < -7 d). The residence time is an additional argument for neglecting the short-lived products from the ^{232}Th decay chain with respect to dose calculation. The mean ^{210}Pb concentration value of 248 ± 145 µBq m^{-3} found in this study (cf. Table 2) is in agreement with data published by the Bundesministerium für Umwelt, Naturschutz und Reaktorsicherheit (BMU – Federal Ministry for the Environment, Nature Conservation and Nuclear Safety)[8] (200-670 µBq m^{-3}) and Gründel et al. [3], who report 100 – 500 µBq m^{-3}.

Applying the parameters of Table 1 to the ^{210}Pb concentration presented in Table 2, the annual dose for children <= 0 year and adults can be calculated. Results are shown in Table 3.

Table 3. Annual inhalation dose for children <= 1 year and adults

	Effective Dose	Lung Dose
Children	8.7 ± 4.8 µSv	72.5 ± 40.2 µSv
Adults	10.5 ± 5.8 µSv	85.9 ± 47.6 µSv

Of course, it must be stated here that the dose values of Table 3 are fictitious because an unrealistic outdoor exposure time of 365 d is assumed here. With respect to the initial problem this investigation focuses on the long-lived activity adsorbed onto aerosols and not on the full range of ^{222}Rn decay products. Moreover, indoor exposure to radon and its decay products is much greater than outdoor exposure, e.g. [6].

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